

DATE ISSUED JUN 27 1978 40

ORNL/TM-6260

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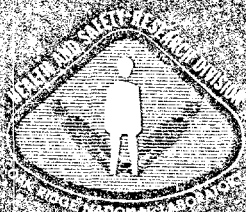
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Assessment of ^{99}Tc Releases
to the Atmosphere—
A Plea for Applied Research

J. E. Till
F. O. Hoffman
D. E. Dunning, Jr.

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Printed in the United States of America. Available from
National Technical Information Service
U.S. Department of Commerce
5285 Port Royal Road, Springfield, Virginia 22161
Price: Printed Copy \$4.50; Microfiche \$3.00

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ORNL/TM-6260
Dist. Categories UC-11, UC-41

Contract No. W-7405-eng-26

Health and Safety Research Division

ASSESSMENT OF ^{99}Tc RELEASES TO THE ATMOSPHERE —
A PLEA FOR APPLIED RESEARCH

J. E. Till¹, F. O. Hoffman², and D. E. Dunning, Jr.

¹Subcontract
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Date Published - June, 1978

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ACKNOWLEDGMENTS

The authors gratefully acknowledge, with thanks, E. L. Etnier of the Oak Ridge National Laboratory, R. G. Gast and L. Thorvig of the University of Minnesota, Y. C. Ng of Lawrence Livermore Laboratory, and J. K. Soldat and R. E. Wildung of Battelle Pacific Northwest Laboratory for their assistance during the preparation of this manuscript.

ABSTRACT

Recent experimental data suggest that the concentration factor for uptake of ^{99}Tc by vegetation from soils may be two to three orders of magnitude higher than the 0.25 value currently being used in radiological assessments. Following a survey of the literature, a concentration factor of 50 was applied to evaluate the dose from a 1.0 Ci/year release to the atmosphere by a hypothetical uranium enrichment facility. Doses to the GI tract and thyroid of an adult living 1600 m from the facility were 18 millirems and 80 millirems, respectively. These doses are delivered entirely through transport of ^{99}Tc through food chain pathways. This assessment indicates a potential for ^{99}Tc exposures to exceed recently proposed standards of the U.S. Environmental Protection Agency in 40 CFR 190. The previously assumed concentration factor of 0.25 would have produced corresponding doses of 0.13 millirem to the GI tract and 0.57 millirem to the thyroid.

The results of this analysis demonstrate the need for additional research on the environmental behavior and dosimetry of ^{99}Tc . In particular, data are needed to elucidate the retention of ^{99}Tc in soils and the uptake of ^{99}Tc by edible vegetation in field studies of chronic exposure conditions. Data on the uptake and retention of ^{99}Tc in humans are also necessary to improve the reliability of dose conversion factors for specific organs and various age groups.

INTRODUCTION

Recent data on vegetation uptake of technetium¹⁻³ indicate that soil-to-plant concentration factors ($\mu\text{Ci/g}$ tissue per $\mu\text{Ci/g}$ soil) for this element may be significantly higher than those currently being used in radiological assessments.⁴ In view of this information, there is a need to reevaluate the contribution of the dose from ^{99}Tc relative to that from other radionuclides produced in and released to the environment by nuclear fuel cycle facilities in order to determine the circumstances, if any, under which ^{99}Tc releases would be significant.

Technetium is one of two elements (the other being promethium) with an atomic number lower than bismuth (83) for which there are no stable isotopes. The yield from thermal neutron fission of ^{235}U for ^{99}Tc is high, 6.2%/fission,⁵ or about 0.84 kg per metric ton of uranium in typical spent pressurized water reactor fuel (compared to 1.3 kg per metric ton of ^{137}Cs and 0.55 kg per metric ton of ^{90}Sr).⁶ Following formation in its elemental state during the fission process, technetium is likely to be converted to pertechnetic acid (HTcO_4) during dissolution at the reprocessing plant. A portion of the technetium follows the uranyl nitrate through the solvent extraction process and is converted to technetium heptaoxide (Tc_2O_7) during the denitration or calcining step.⁷ However, routine releases of ^{99}Tc to the atmosphere at a typical commercial fuel reprocessing plant are projected to be only about 45 μCi each year.⁸ Because the resulting doses to individuals living in the vicinity of the reprocessing plant are not expected to be significant relative to the doses received from the release of the other nuclides, ^{99}Tc is ordinarily omitted from the source term.

In the UF_6 conversion process, technetium reacts with fluoride to form volatile compounds that are carried through the gaseous diffusion enrichment step. Small amounts of the technetium may then be released to the environment via gaseous and liquid effluent streams. Both measured and calculated releases to the atmosphere from enrichment plants have been reported with values ranging between 0.54 Ci/year and 5.9 Ci/year (refs. 9 and 10, respectively).

It is important to note that the predominant chemical forms of ^{99}Tc released to the environment have not been determined. However, Wildung et al.³ have stated that the most stable chemical species in aqueous solution is the pertechnetate ion (TcO_4^-), and it is this form which is most likely to enter surface soils. It should also be noted that several techniques have been developed for effectively removing substantial amounts of ^{99}Tc from UF_6 feed material.¹¹ Therefore, if site-specific investigations were to reveal that the environmental levels of ^{99}Tc in the vicinity of nuclear facilities exceed regulatory standards, selective removal of ^{99}Tc from recycled fuel could be implemented.

UPTAKE OF TECHNETIUM BY PLANTS — A REVIEW OF AVAILABLE DATA

Until recently, virtually no data were available in the literature on the uptake of technetium by plants or the behavior of technetium compounds in soils. This lack of information is partly due to the fact that low levels of naturally occurring technetium isotopes are difficult to detect, and environmental contamination from weapons

fallout has received only limited attention. In lieu of information about technetium in soils and vegetation, Ng et al.¹² relied upon a series of assumptions which related the behavior of technetium in the environment to that of iodine, enabling them to derive a soil-to-plant concentration factor of 0.25 ($\mu\text{Ci/g}$ fresh weight tissue per $\mu\text{Ci/g}$ dry weight soil). This is the assumed relationship that has been adopted throughout the assessment literature for technetium in soils and vegetation.^{4,13,14}

In 1974, Gast¹⁵ began to study ^{99}Tc absorption by soils and plant uptake. Plants grown in soils watered with a solution containing ^{99}Tc pertechnetate (TcO_4^-) showed appreciable uptake and translocation into the above ground tissue with concentrations in seeds being much less than concentrations in vegetative tissue. The observed soil-to-plant concentration factors were as much as three orders of magnitude greater than the assumed 0.25 (Table 1). However, in these laboratory experiments no effort was made to achieve an equilibrium level of ^{99}Tc between soil and plant tissue. Also, the plants in these experiments did not reach maturity prior to sampling. Furthermore, the concentration at which the ^{99}Tc was applied was significantly higher than what one would expect to find in the environment around a nuclear installation. Cataldo et al.¹⁶ have reported that the uptake of technetium may be affected by the concentration of technetium in soil, with a greater rate of uptake at higher concentrations. These laboratory data, therefore, probably do not provide an accurate estimate of uptake by plants in the environment around a nuclear facility where soils would

Table 1. Concentration factors for ^{99}Tc based upon
soil to aerial parts of wheat seedlings
($\mu\text{Ci/g}$ wet weight of plant per $\mu\text{Ci/g}$
dry weight of soil)^a

Type of soil	Concentration factor			
	Soils watered with solution containing ^{99}Tc		Soils moist incubated with ^{99}Tc	
	<u>Unfertilized</u>	<u>Fertilized</u>	<u>Unfertilized</u>	<u>Fertilized</u>
Bearden	166	83	165	18
Hegne	213	137	128	32
Hibbing	185	129	253	56
Nicollet (surface)	228	158	268	127
Nicollet (subsurface)	213	209	249	326
Omega	124	76	53	40
Bergland	191	144	130	86
Arveson	130	48	89	91
Waukegan	209	158	269	189
Zimmerman	200	149	153	107
(Sapric peat)	106	112	143	54

^aDerived from the experiments by Gast et al.¹ by using conversion factors
for dry to wet weight provided by Thorvig.¹⁸

be subject to continuous contamination as a result of routine releases of ^{99}Tc .

Soil-to-leaf concentration factors for wheat and soybean plants that were derived from data published by Wildung et al.^{2,17} are listed in Table 2. As with the experiments reported by Gast,¹ the level of ^{99}Tc in laboratory soil was not maintained constant for the duration of the test, and therefore, concentration factors listed in Table 2 may not be representative of what would be expected under equilibrium conditions.

More recent data have been obtained on ^{99}Tc uptake¹⁸ in which wheat plants were germinated and grown for 10 days in 1900 ml of ^{99}Tc labeled one-third strength Hoagland mixture. In this solution the concentration of ^{99}Tc was maintained at a constant level ($\pm 10\%$) and the resulting solution-plant concentration factors were approximately 50 ($\mu\text{Ci/g}$ wet weight of tissue per $\mu\text{Ci/g}$ solution). This value could be interpreted to represent a potential "upper limit" for the soil-to-plant concentration factor because the ^{99}Tc in solution should be more available for plant uptake via the roots than ^{99}Tc in soil. However, contrary to this expectation, Cataldo et al.¹⁶ have demonstrated that pertechnetate applied to soils is readily available for plant uptake. In addition, the factor of 50 derived from the Hoagland mixture experiments is comparable to the concentration factors determined by Wildung et al. in Table 2 in which a state of equilibrium was not attained.

The choice of a concentration factor for use in environmental assessments should be based on experiments representative of field

Table 2. Concentration factors for ^{99}Tc based upon soil to aerial parts of soybean and wheat ($\mu\text{Ci/g}$ wet weight of plant tissue per $\mu\text{Ci/g}$ dry weight soil)^a

Concentration of Tc ($\mu\text{g Tc/g soil}$)	Concentration factor	
	Soybeans	Wheat
0.01	35	38
0.1	17	43
1.0	95	no growth
5.0	19	no growth

^aDerived from the experiments by Wildung et al.^{2,17}

conditions when a state of equilibrium has been obtained between soil and plant. An estimate of the upper range of the soil-to-plant concentration factor for ^{99}Tc is precluded because of the fact that no measurements have been performed under equilibrium conditions and because there are not stable technetium isotopes in the environment from which to derive this information.

It is of interest to note that an absolute maximum soil-to-plant concentration factor for ^{99}Tc (or any other isotope for which specific data were not available) was assumed to be 100 (pCi/g dry weight tissue per pCi/g dry weight soil) by Ng. et al.¹² Using the value for moisture content in vegetation of 75% assumed by Ng et al., this concentration factor converts to a value of 25 (pCi/g wet weight tissue per pCi/g dry weight soil). This assumption was made in order to include in their comprehensive handbook conservative values for ^{99}Tc concentrations in beef and in the total body of man. Nevertheless, this value of 25 is significantly less than the values reported in Table 1.

Because of the incomplete nature of the data currently available, we feel that it is not possible to derive a "best estimate" of the soil-to-plant concentration factor for ^{99}Tc . In fact, the values in Table 1 would seem to indicate that a concentration factor of 50 would be among the lowest to be assumed for calculational purposes, especially since these values are not representative of equilibrium conditions. However, our analysis of reported concentration factors for ^{99}Tc in soil and plants suggests that the value presently being used, 0.25, is a significant underestimate of uptake of ^{99}Tc through the roots. On the other hand, it is entirely possible that the concentration factors obtained under laboratory conditions could be an overestimate. Nevertheless, one

cannot be justified at this time in disregarding the data shown in Tables 1 and 2 and continuing to assume a concentration factor of 0.25. Despite the uncertainties associated with the choice of a value for soil-to-plant uptake of ^{99}Tc , we will assume a value of 50 for this study to determine the significance of using elevated values for this concentration factor in radiological assessment models.

DOSE CONVERSION FACTORS

Dose estimates used in this study were calculated with the INREM-II computer code.¹⁹ The INREM-II code employs an adaptation of the ICRP Task Group Lung Model²⁰ and a catenary GI tract model based on the transit times of Eve²¹ to describe the dynamics of retention in the respiratory and gastrointestinal tracts, respectively. Retention in other systemic organs is represented by a series of decaying exponential terms. For all inhalation calculations performed in this study, the Task Group Lung Model solubility category D is assumed.

Knowledge of the chemical form of technetium is crucial to assessing its distribution in the body. Pertechnetate ion (TcO_4^-) concentrates in the thyroid, salivary glands, kidneys, and gastrointestinal tract.²²⁻²⁵ Aggregated and colloidal forms of technetium are localized in the liver.^{23,26} Numerous other chemical complexes of the element have specific clinical applications due to their characteristic localization in particular tissues. In this report, however, we consider only the pertechnetate ion which appears to be the most likely form prevailing in the environment.^{1,3}

Table 3 presents estimates of dose conversion factors in millirems per pCi intake of ^{99}Tc pertechnetate based upon three different sets of metabolic assumptions for a reference adult. The dose conversion factors in set (A) of Table 3 have been derived from basic metabolic information given by the International Commission on Radiological Protection.²⁷ Biological retention of technetium is described by a single exponential term for each organ of concern.

From an analysis of $^{99\text{m}}\text{Tc}$ dosimetry performed in WASH-1400 (ref. 28) the Nuclear Regulatory Commission estimates a uniform distribution throughout the body with the exception of the thyroid; for the thyroid a fractional uptake of 0.54% from blood is specified. Biological retention in all tissues is described with a multicompartment exponential model of Beasley et al.²² A fractional uptake of 95% from the small intestine to blood is assumed with only 5% of the initial GI tract contents of technetium reaching the critical lower large intestine. The second set of dose conversion factors (B) in Table 3 is computed from these data.

Other studies have indicated a rapid initial pertechnetate uptake of approximately 2% to the thyroid from blood with some estimates as high as 8 to 10% in euthyroid²⁴ and 6 to 30% in hyperthyroid individuals.²³ From this range of values, 2% has been adopted in the third set of dose conversion factors (C) in Table 3. While not entirely arbitrary, this selection of an appropriate uptake factor is an additional uncertainty in the calculations. A fractional uptake of 80% is assumed from the small intestine to blood.²⁹ Thus 20% of the technetium in the GI tract reaches the lower large intestine. Biological retention of ^{99}Tc in

Table 3. Adult dose conversion factors^a for ⁹⁹Tc pertechnetate (millirems/pCi)

Set	Pathway	Total body	Bone	Kidneys	GI tract ^b	Thyroid
A ^c	Ingestion	5.0 E-8	1.3 E-7	2.3 E-6	6.1 E-6	ND ^d
	Inhalation	1.3 E-8	3.1 E-8	5.8 E-7	7.5 E-6	ND
B ^e	Ingestion	2.5 E-7	4.1 E-7	4.1 E-7	8.0 E-7	4.6 E-6
	Inhalation	1.8 E-7	2.7 E-7	2.7 E-7	1.3 E-7	3.0 E-6
C ^f	Ingestion	2.1 E-7	3.6 E-7	4.6 E-7	3.2 E-6	1.4 E-5
	Inhalation	1.7 E-7	2.7 E-7	3.5 E-7	5.1 E-7	1.1 E-5

^aDose conversion factor as used in this paper is the 50-year integrated dose resulting from a unit intake of ⁹⁹Tc.

^bDose conversion factors in (B) and (C) are calculated for the lower large intestine, which is the critical segment of the GI tract.

^cSee ref. 31. Set (A) was derived from basic dosimetric data given by the International Commission on Radiological Protection.²⁷

^dND = No data reported for the thyroid.

^eSet (B) represents a calculation based on the dosimetric data in WASH-1400 (ref. 28), using the INREM-II computer code.¹⁹

^fSet (C) was calculated with the INREM-II methodology and data base.¹⁹

organs other than the lungs and GI tract is estimated with the model of Beasley et al.²²

The metabolic model presented by Beasley et al.²² and used in parts (B) and (C) of Table 3 is based upon a clinical study of the short-lived ^{95m}Tc and ^{96}Tc isotopes in man. Studies with ^{99m}Tc indicate similar patterns of pertechnetate uptake and retention by the thyroid, kidneys, and GI tract, although sometimes suggesting more rapid clearance from these tissues^{25,30} and erratic rates of uptake.²⁹ One might be somewhat skeptical about applying this metabolic model to the long-lived ^{99}Tc isotope since studies of these short-lived isotopes may not provide adequate information concerning possible long-term retention in tissues of interest. Clinical studies indicate a rapid decrease of pertechnetate concentrations in the thyroid,²⁶ but these are based upon a single administration to the patient. Additionally, it is recognized that the retention model was based upon total-body elimination of technetium and would not necessarily be representative of retention in individual organs. However, in lieu of organ-specific data, the Beasley retention model has been used with the INREM-II computer code to estimate dose conversion factors in our analysis.

The dose to specific organs could be greater for a child than for an adult under identical conditions of exposure. When only the difference in organ mass is considered, the dose conversion factor is elevated one order of magnitude for ingestion and inhalation.³¹ Other physiological and extrinsic age-dependent differences might also influence uptake and retention of ^{99}Tc . For example, differences in

dietary habits would make milk a more prominent component of the child's diet than that of the adult. It is evident that further studies are needed to specify dose conversion factors applicable to critical groups of the population. For the purposes of this paper we will use the dose conversion factors listed in Table 3 (C), which are applicable to a reference adult.

ASSESSMENT OF THE DOSE FROM TECHNETIUM RELEASED TO THE ENVIRONMENT

In order to determine the significance of technetium releases to the environment, we assumed for the purposes of an initial analysis a soil-to-plant (fresh weight) concentration factor of 50 for ^{99}Tc in crops and calculated the dose to an individual living 1600 m from a hypothetical gaseous diffusion enrichment plant handling recycled uranium. Meteorological data for this analysis were taken from a report by Roddy et al.³² The parameters used to estimate the atmospheric transport of technetium released to the environment are listed in Table 4. The AIRDOS-II computer code³³ was used to calculate the air concentration and ground deposition rate of ^{99}Tc at the point of exposure.

The concentration of ^{99}Tc in vegetables, meat, and milk was computed following procedures outlined in the U.S. Nuclear Regulatory Commission Guide 1.109 (ref. 4) and using a revised transfer coefficient for ^{99}Tc in milk of 9.9×10^{-3} day/liter (ref. 34) and in meat of 8.7×10^{-3}

Table 4. Parameters used to estimate the
atmospheric transport of technetium
released to the environment

<u>Parameter</u>	<u>Value</u>
Release rate	1 Ci/year
Release height	20 m
χ/Q at 1600 m	$9.9 \times 10^{-7} \text{ sec/m}^3$
Annual average deposition velocity for wet and dry processes	$1.1 \times 10^{-2} \text{ m/sec}$
Deposition rate at 1600 m	$3.02 \times 10^1 \text{ pCi/m}^2 \text{ day}$

day/kg (ref. 12). Results of the calculations are listed in Table 5 for soil-to-plant concentration factors of 0.25 (the value most commonly assumed in earlier assessments) and 50 (the value assumed in this analysis). Direct deposition onto above ground vegetation contributes approximately 20% of the ^{99}Tc concentration in vegetables, meat, and milk when the factor of 0.25 is employed but proves negligible when the factor 50 is considered. Corresponding ^{99}Tc ingestion rates for a reference adult individual based upon the concentrations listed in Table 5 are shown in Table 6. It is evident from Table 6 that each of the ingestion pathways contributes significantly to the total technetium intake.

The dose commitments to a reference adult residing 1600 m from the point of release and obtaining his entire food supply from that location are presented in Table 7. These doses have been calculated using the dose conversion factors in Table 3 (C) and the intake rates listed in Table 6. According to our calculations, the organs receiving the highest dose are the thyroid and the GI tract. Increasing the soil-to-plant concentration factor from 0.25 to 50 results in an increase in the dose commitment via the ingestion pathway of approximately 140 times. The assumption of a value of 50 for the concentration factor produces a dose commitment to the thyroid of 80 millirems and a dose commitment of 18 millirems to the GI tract. The use of 0.25 as the concentration factor produces a dose commitment of 0.57 millirems to the thyroid and 0.13 millirem to the GI tract. The inhalation pathway does not contribute significantly to the dose commitment in either case.

Table 5. Concentration of ^{99}Tc in vegetables, meat, and milk 1600 m from a gaseous diffusion enrichment plant releasing 1 Ci/year to the atmosphere^a

Soil-to-plant concentration factor for ^{99}Tc	^{99}Tc concentration in:		
	Vegetables (pCi/kg)	Meat (pCi/kg)	Milk (pCi/liter)
0.25	2.3×10^2 ^b	1.1×10^2 ^b	1.3×10^2 ^b
50	3.4×10^4 ^c	1.5×10^4 ^c	1.7×10^4 ^c

^a Assuming 15 years of accumulation in soil.⁴

^b Approximately 20% of the ^{99}Tc was contributed via direct deposition onto above ground vegetation.

^c Contribution from direct deposition is insignificant.

Table 6. Intake rate of ^{99}Tc by an adult living 1600 m from a gaseous diffusion enrichment plant releasing 1 Ci/year to the atmosphere

Soil-to-plant concentration factor for ^{99}Tc	Intake rate (pCi/year) via ingestion of			
	Vegetables ^a	Meat ^b	Milk ^c	Total
0.25	1.5×10^4	1.4×10^4	1.2×10^4	4.1×10^4
50	2.2×10^6	1.6×10^6	1.9×10^6	5.7×10^6

^a Assuming 175 g of vegetables ingested daily, based on data from the International Commission on Radiological Protection, Report of the Task Group on Reference Man, ICRP-Publication 23, Pergamon Press, Oxford, 1975.

^b Assuming 300 g of meat ingested daily.⁴

^c Assuming 0.3 liter of milk ingested daily, based on data from the International Commission on Radiological Protection, Report of the Task Group on Reference Man, ICRP-Publication 23, Pergamon Press, Oxford, 1975.

Table 7. Maximum individual 50-year dose commitment from ^{99}Tc to an adult living 1600 m from the stack of a gaseous diffusion plant releasing 1 Ci/year to the atmosphere

Organs	<u>50-year dose commitment^a (millirem)</u>				
	Total body	Bone	Kidneys	GI tract	Thyroid
Concentration factor	50	0.25	50	0.25	50
Ingestion dose	1.2	8.6×10^{-3}	1.8	1.3×10^{-2}	18
Inhalation dose	4.4×10^{-5}	7.0×10^{-5}	9.1×10^{-5}	1.3×10^{-5}	2.9×10^{-3}

^aDose commitment as used in this paper is the 50-year integrated dose resulting from a chronic intake of ^{99}Tc over one year.

DISCUSSION

It is important to note that in this assessment, several assumptions have been made which could increase or decrease the resulting dose calculations: for example, the specification of a diet entirely produced and consumed locally. In reality, local agricultural production typically would be supplemented by foods imported from outside the assessment area. This assumption would therefore cause an overestimate of the dose to the average individual.

Another assumption that would overestimate the dose is the effective time of accumulation of ^{99}Tc as pertechnetate in the root zone of the soil. Because of its apparent high solubility, and subsequent high mobility in soil,³ the soil accumulation time for pertechnetate may be less than the 15 years assumed in Regulatory Guide 1.109 (ref. 4) and in this study. The Regulatory Guide assumes that the only loss of a given isotope in the soil root zone is the loss due to radioactive decay. This assumption leads to substantial build-up of ^{99}Tc in soils during the lifetime of a nuclear facility. An additional mechanism for removing ^{99}Tc from soils would be the successive harvesting of vegetation which exhibit high soil-to-plant concentration factors for this isotope. Therefore, the assumed 15-year soil accumulation time used in this study may be unrealistic.

On the other hand, our assumed source term for an enrichment facility of 1 Ci/year is significantly lower than several values already reported in the literature (e.g., 5.9 Ci/year, ref. 10). Also, the location of exposure in this study, 1600 m from the source was not the point at which the maximum air concentration would be

expected to occur. Our calculations indicate that the maximum ground level air concentration to be considered for assessment purposes occurs at 800 m, where the deposition rate would be 7.0×10^{-4} pCi/m²-sec. We selected the 1600 m distance because it is more representative of sites for enrichment and reprocessing facilities presently in existence.

One additional lack of conservatism in this study is the use of a reference adult as the target individual. Consideration of dose conversion factors and dietary habits representative of children could result in a higher estimate of dose. A reference adult was used because of the relative absence of dosimetric data applicable to other age groups.

The significance of the dose commitments resulting from a one curie per year release of ⁹⁹Tc to the atmosphere can be put into perspective by comparing them to recent radiation protection standards promulgated by the U.S. Environmental Protection Agency in 40 CFR 190 (ref. 35). These standards recommend 25 millirems per year as the maximum dose to the total body and other organs except the thyroid; the standard for the thyroid is 75 millirems per year. For a one curie per year release to the atmosphere and a soil-to-plant concentration factor of 50, our analysis indicates that doses to the total body, bone, and kidneys would be less than the EPA standards. The dose to the GI tract would approach, and the dose to the thyroid would be slightly greater than these standards. However, even a minor change in the calculational assumptions, as described earlier, could increase the GI tract dose enough to exceed the standard.

It is important to emphasize that we do not intend to imply that a problem necessarily exists with ⁹⁹Tc releases to the environment on the basis of these tentative calculations. What we do stress, however, is

that additional research is urgently needed. Environmental assessments are presently bound to employ calculations using an elevated value for the soil-to-plant transfer of ^{99}Tc without sufficient data being available to refine other parameters.

CONCLUSIONS

It is concluded from this analysis that there is an urgent need for environmental monitoring and additional experimental data, particularly data derived under field conditions describing ^{99}Tc uptake of vegetation and mobility in soils. Data must be obtained from concentrations of ^{99}Tc in soil representative of levels to be expected from routine releases from nuclear facilities. Concentration factors should be derived under equilibrium conditions and specified for the edible portions of plant tissue. In addition, data must be obtained on the physicochemical form of release of ^{99}Tc to the atmosphere and its environmental transport properties.

Significant uncertainties exist in the uptake and retention of technetium in man and the subsequent specification of dose conversion factors for specific age groups and individual organs. Little information is available to provide insight into possible long-term retention of ^{99}Tc in organs of interest (notably the thyroid); such data are needed to accurately assess the dose due to chronic exposure to routine releases from a nuclear facility. There is an urgent need for information describing the uptake and retention of ^{99}Tc in children, since they could comprise the critical segment of the population at risk.

This assessment indicates that ^{99}Tc is a potentially critical radionuclide in the nuclear fuel cycle. However, the confirmation of this assessment can only be determined after improving the state of knowledge about the environmental behavior and dosimetric properties of this isotope.

REFERENCES

1. R. G. Gast, E. R. Landa, and L. J. Thorvig. The Behavior of Technetium-99 in Soils and Plants, Progress Report for the Period April 1, 1974-March 31, 1977, USERDA Contract No. Ey-76-5-20-2447, COO-2447-5 (1976).
2. R. E. Wildung, T. R. Garland, and D. A. Cataldo. Accumulation of Technetium from Soil by Plants - A Potential Mechanism for Uptake and Toxicity, Battelle-Northwest Laboratory, BNWL-SA-5393 (1975).
3. R. E. Wildung, T. R. Garland, and D. A. Cataldo, Health Phys. 32, 4 pp. 315-317 (1977).
4. U.S. Nuclear Regulatory Commission. Calculation of Annual Doses to Man from Routine Releases of Reactor Effluents for the Purpose of Evaluating Compliance with 10 CFR Part 50, Appendix I, Regulatory Guide 1.109 (1976).
5. R. D. Evans. The Atomic Nucleus, McGraw-Hill, New York (1970).
6. B. C. Finney, R. E. Blanco, R. C. Dahlman, G. S. Hill, F. G. Kitts, R. E. Moore, and J. P. Witherspoon. Correlation of Radioactive Waste Treatment Costs and the Environmental Impact of Waste Effluents in the Nuclear Fuel Cycle-Reprocessing Light-Water Reactor Fuel, Oak Ridge National Laboratory ORNL/NUREG/TM-6 (1976).

REFERENCES (CONT'D)

7. A. J. Story. Letter to Mr. W. R. Golliher, Union Carbide Corporation, Subj. "Status Report on the Behavior of Tc and NO_xUF_x Compounds in the Cascade as of October 28, 1974."
8. B. C. Finney. Oak Ridge National Laboratory, personal communication, 1977.
9. U.S. Energy Research and Development Administration, Final Environmental Statement, U.S. Nuclear Power Export Activities, ERDA-1542 (1976).
10. U.S. Energy Research and Development Administration, Environmental Statement, Portsmouth Gaseous Diffusion Plant Expansion, ERDA-1549, Vol. 1 (1977).
11. A. J. Story. Union Carbide Corporation, Paducah, Ky., personal communication, 1977.
12. Y. C. Ng, C. S. Burton, S. E. Thompson, R. K. Tandy, H. K. Kretner, and M. W. Pratt. Prediction of the Maximum Dose to Man from the Fallout of Nuclear Devices, Handbook for Estimating Internal Dose from Radionuclides Released to the Biosphere, University of California-Lawrence Livermore Laboratory, UCRL-50163, part IV (1968).
13. G. G. Killough and L. R. McKay. A Methodology for Calculating Radiation Doses from Radioactivity Released to the Environment, Oak Ridge National Laboratory Report ORNL-4992 (1976).

REFERENCES (CONT'D)

14. D. A. Baker, G. R. Hoenes, and J. K. Soldat. FOOD-An Interactive Code to Calculate Internal Radiation Doses from Contaminated Food Products, Battelle-Northwest Laboratory, BNWL-SA-5523 (1976).
15. R. G. Gast. The Behavior of Technetium-99 in Soils and Plants, Progress Report for the Period April 1, 1974-March 31, 1975, USAEC Contract No. AT(11-1)-2447, COO-2447-1 (1975).
16. D. A. Cataldo, R. E. Wildung, and T. R. Garland. "Technetium Accumulation, Fate and Behavior in Plants," in Proc. Symp. on the Environmental Chemistry and Cycling Processes, Augusta, Georgia, April 27-30, 1976.
17. R. E. Wildung. Battelle-Northwest Laboratory, personal communication, 1977.
18. L. Thorvig. University of Minnesota, personal communication, 1977.
- 19.. G. G. Killough, J. C. Pleasant, and D. E. Dunning. INREM-II: A Computer Implementation of Recent Models for Estimating the Dose Equivalent to Organs of Reference Man from an Inhaled or Ingested Radionuclide, Oak Ridge National Laboratory Report ORNL/NUREG/TM-84.
20. P. E. Morrow, D. V. Bates, B. R. Fish, T. F. Hatch and T. T. Mercer. Health Phys. 12, 173-207 (1966).
21. I. S. Eve. Health Phys. 12 131-162 (1966).

REFERENCES (CONT'D)

22. T. M. Beasley, H. E. Palmer, and W. B. Nelp. Health Phys. 12, 1425 (1966).
23. P. V. Harper, K. A. Lathrop, R. J. McCardle, and G. Andros. "The Use of Technetium-99m as a Clinical Scanning Agent for Thyroid, Liver and Brain," Medical Radioisotopes Scanning, Vol. II, International Atomic Energy Agency, 1964.
24. I. Kazem, P. Gelinsky, and P. Schenck. Br. J. Radiol. 40, 292 (1967).
25. E. M. Smith. J. Nucl. Med. 6, 231 (1965).
26. J. G. McAfee, C. F. Fueger, H. S. Stern, H. N. Wagem, Jr., and T. Migita. J. Nucl. Med. 5, 811 (1964).
27. Recommendations of the International Commission on Radiological Protection, ICRP Publication 2, Pergamon Press, Oxford, 1959.
28. U.S. Nuclear Regulatory Commission. Reactor Safety Study: An Assessment of Accident Risks in U.S. Commercial Nuclear Power Plants, Appendix VI. Calculation of Reactor Accident Consequences WASH-1400. PB-248206, NUREG 75/104 (1975).
29. M. T. Hays and B. Wesselosky. J. Nucl. Med. 14, 331 (1973).
30. M. T. Hays. J. Nucl. Med. 14, 331 (1973).
31. G. R. Hoenes and J. K. Soldat. Age-Specific Radiation Dose Commitment Factors for a One-year Chronic Intake, Nuclear Regulatory Commission Report, NUREG-1072 (1977).

REFERENCES (CONT'D)

32. J. W. Roddy, R. E. Blanco, G. S. Hill, R. E. Moore, R. D. Seagren, and J. P. Witherspoon. Correlation of Radioactive Waste Treatment Costs and the Environmental Impact of Waste Effluents in the Nuclear Fuel Cycle-Fabrication of High-Temperature Gas-Cooled Reactor Fuel Containing Uranium-233 and Thorium, Oak Ridge National Laboratory Report ORNL/NUREG/TM-5 (1976).
33. R. E. Moore. The AIRDOS-II Computer Code for Estimating Radiation Dose to Man from Airborne Radionuclides in Areas Surrounding Nuclear Facilities, Oak Ridge National Laboratory Report ORNL-5245 (1977).
34. Y. C. Ng, C. S. Colsher, D. J. Quinn and S. E. Thompson. Transfer Coefficients for the Prediction of the Dose to Man via the Forage-Cow-Milk Pathway from Radionuclides Released to the Biosphere, University of California-Lawrence Livermore Laboratory, UCRL-51939 (1977).
35. U.S. Environmental Protection Agency. Environmental Radiation Protection Requirements for Normal Operations of Activities in the Uranium Fuel Cycle, 40 CFR 190, EPA 520/4-76-016 (1976).

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